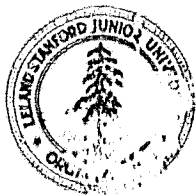


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**Department of AERONAUTICS and ASTRONAUTICS  
STANFORD UNIVERSITY**

**Final Report**

**July 1, 1980 - June 30, 1982**

**on**

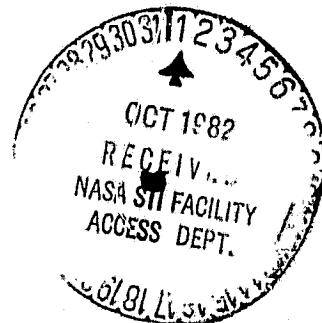
**DENSITY MEASUREMENT IN AIR WITH A SATURABLE ABSORBING SEED GAS**

**NASA-Ames Grant No. NAG 2-36**

**Submitted to the  
National Aeronautics and Space Administration  
Ames Research Center  
Moffett Field, California 94035  
R.L. McKenzie - Technical Officer**

**by the**

**Department of Aeronautics and Astronautics  
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Stanford, California**



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**Principal Investigator**

**✓ Donald Baganoff**

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## ABSTRACT

Several approaches having potential for making density measurements in a compressible flow, where one or more laser beams are used as probes, are investigated. Saturation in sulfur hexafluoride and iodine are considered together with a crossed-beam technique where one beam acts as a saturating beam and the other is at low intensity and acts as a probe beam. The cross-coupling phenomena is present in both gases and is quite pronounced at low pressures, but because a high degree of saturation is not found in practice for a variety of reasons, especially at higher pressures where application would be most desired, the method is not fully pursued.

It is shown that a balance between an increase in fluorescence intensity with increasing pressure owing to line broadening and the normal decrease in intensity with increasing pressure owing to quenching can be used to develop a linear relation between fluorescence intensity and number density and thereby lead to a new density measurement scheme. The effect is found in iodine fluorescence when iodine is excited by the line-narrowed output of an argon-ion laser detuned from an iodine absorption line by 3 GHz. The method is used to obtain a density image of the cross-section of an iodine-seeded underexpanded supersonic jet of nitrogen, by illuminating the cross-section by a sheet of laser light.

## REVIEW OF PROGRESS

The overall objective of the work has been to develop a method whereby one is able to make a measurement of gas density at a point in a non-steady, three-dimensional, compressible flow, using one or more laser beams as probes. The work covered a period of two years (July 1980 - June 1982) and several approaches having good potential were studied in the period. Our attention was initially focused on the saturation properties of sulfur hexafluoride and the interaction that develops when one crosses two  $\text{CO}_2$  laser beams, of sufficient intensity, in a gas seeded with sulfur hexafluoride. This work formed the basis of our proposal and early results were reported at an American Physical Society meeting on fluid dynamics (see section on presentations). Because of the difficulty of working with infrared optics and detectors, we then shifted our attention to the use of iodine and the argon-ion laser and extended our scope of research to include fluorescence. Most of our work was done with the iodine molecule and the argon-ion laser. The principal publications which have resulted from this work are a Ph.D. thesis by J.C. McDaniel and a paper in the Physics of Fluids (see section on publications).

### Sulfur Hexafluoride

The properties of sulfur hexafluoride that make it a potential candidate for use in a density measurement scheme are: the coincidence of its absorption band with the emission band of the  $\text{CO}_2$  laser; it is a nontoxic substance which allows it to be used in a facility which is open to a laboratory environment; and it does not degrade materials that it comes into contact with, and therefore most any material may be used in the construction of a test facility which employs it. The concept that we

developed is an extension of the idea on which absorption tomography is based. Absorption tomography makes use of a constant absorption coefficient, where absorption is proportional to gas density and independent of laser beam intensity, and in order to reconstruct the density at a point, one needs, in effect, an infinite number of beams passing through the point at which the density is to be determined. On the otherhand, if a nonlinear medium is present, the number of crossing beams can be reduced to two, which greatly reduces the complexity of the experimental setup and the corresponding data reduction effort required.

The nonlinear situation we studied was saturation in sulfur hexafluoride. In a saturable medium, the absorption coefficient is a function of laser intensity. Consequently, if one uses a combination of a weak probe beam and a strong saturating beam, one may modulate the transmission of the probe beam through a saturable medium by crossing it with the strong saturating beam which is being turned off and on. The modulated amplitude of the probe beam gives quantitative information on the interaction at the crossing point, and allows a determination of the gas density at the crossing point through the known nonlinear properties of the medium. The attractiveness of the scheme is that the detector that monitors the probe beam intensity lies outside of the medium and the measurement is effectively made at a point in space defined by the crossing point of the two beams.

In our experiments we used a 10 Watt CW  $\text{CO}_2$  laser and were able to demonstrate the interaction effect with a mixture of only 1 torr sulfur hexafluoride in 760 torr of air. A very interesting response was obtained at low mixture pressures where the saturation intensity is low, because of small excited-state quenching rates, but the effect was less pronounced and

more complicated at mixture pressures near one atmosphere where most applications are desired. One of the major unwanted effects at the higher mixture pressures was found to be a thermal effect which simulated saturation but exhibited the long time constant associated with heat conduction in a gas. In effect, the laser was heating the gas mixture and the higher gas temperature caused a change in the absorption coefficient which dominated the change due to saturation for the level of saturation which could be obtained at the higher pressures with the CW laser used.

The number of nonlinear effects that have potential application to the measurement of density through a crossed-beam technique is very large and ranges from the method we first explored to more recent developments that fall in the category generally called four-wave mixing. In each of these cases the demand on optical components, detectors and optical alignment is quite high and difficult to carry out when using an infrared beam. Because our work was focused on exploring the application of new principles and was not confined to the use of a particular gas, we shifted our attention to the use of iodine and the argon-ion laser, which permitted the work to be carried out in the visible part of the spectrum.

### Iodine

The properties of iodine that make it an attractive candidate are that it strongly absorbs the 514.5 nm output of the argon-ion laser, it has a rather low value for the saturation intensity, and its fluorescence intensity is quite strong, even at pressures near one atmosphere. On the otherhand, it also possesses some undesirable properties that make it hard to work with: it reacts with aluminum and low carbon steel, which requires the use of a suitable coating on all component parts of a facility using

It, and it is sufficiently toxic in concentrations at which it would be used to require its use in a closed test apparatus. Nevertheless, in an exploratory investigation where a general concept is being studied, the advantages are sufficient to warrant its use. Once the technique is developed with iodine, other substances and other laser wavelengths could be used in an actual application.

A detailed study of the saturation properties of iodine were carried out at an iodine concentration equal to its vapor pressure at room temperature and for a range of nitrogen background pressures of up to one atmosphere. In addition, a theoretical model was developed treating the iodine molecule as a three-level system, where one of the levels represented the dissociated state. The theoretical work also considered broadband as well as narrowband excitation together with the effects of nonuniform laser intensity across the beam width on the saturation characteristics of the medium. Most of the work was directed towards understanding the saturation properties of the iodine-nitrogen mixture for the pressure range from 50 torr to atmospheric, which is a considerably higher range of pressures than one finds in earlier studies of iodine.

The mathematical model was able to predict the experimental behavior very well and it provided a solid foundation on which to base our understanding of the saturation process. The main conclusions drawn were as follows. Although the saturation intensity of iodine is low compared with other molecules, complete saturation is never achieved for a variety of practical reasons, even at power levels of several MW/cm<sup>2</sup>. The principal factor causing this result is the nonuniform intensity distribution across the laser beam which always introduces a region of low-intensity light at the edge of the beam, and therefore, a region of unsaturated gas. Shaping



the laser beam intensity into a top-hat profile helps but not if the beam must be focused tightly for which the Gaussian profile again results. Another factor is introduced by the shape of the absorption band of the gas, that is, in the wings of the absorption band the gas is always optically thin and therefore it does not lead to saturation. Partial saturation was obtained for a variety of conditions and for those cases where partial saturation is sufficient, iodine is a good candidate gas. In our case a more complete saturation was desired in order to simplify the relation between the detected interaction signal and the iodine concentration as well as to maximize the signal strength.

The theory developed was also used to analyze the fluorescence intensity data obtained for various test conditions. Our findings here were quite remarkable and led to some new results. When the fluorescence studies were conducted using narrow-band excitation (with an etalon placed in the laser cavity to select a single transition), the shape of the fluorescent-intensity signal with nitrogen pressure changed dramatically as the laser was detuned (up to 3 GHz). The characteristic roll off with pressure caused by quenching was reduced in some cases and flattened in others. A detailed analysis of the theory showed that the behavior was a result of a balancing between two opposing trends: 1) an increase in the fluorescence intensity with increasing pressure owing to line broadening; and 2) the normal decrease in intensity with increasing pressure owing to quenching. When properly adjusted, the relation between fluorescence intensity and number density becomes more direct than for the normal case of quenching, and the method becomes useful as a density measurement technique. This effect was studied extensively and formed the basis of McDaniel's Ph.D. thesis and was the subject of a paper published on the work.

## PUBLICATIONS

1. J.C. McDaniel, "Investigation of Laser-Induced Iodine Fluorescence for the Measurement of Density in Compressible flows", Ph.D. Thesis, Stanford University, Stanford, CA, Dec. 1981. Also published as Stanford University Department of Aeronautics and Astronautics Report (SUDAAR) No. 532, Jan. 1982.

### Abstract

An experimental technique is needed for the nonintrusive measurement of the molecular number density at a point in a compressible flowfield. Laser-induced fluorescence (LIF) is an attractive approach but due to the complication of collisional quenching does not produce a signal that is directly related to the number density. The objective of this work is to investigate the use of LIF for the quantitative measurement of molecular number density in compressible flows. Two approaches for minimizing the quenching effect are explored: saturation and frequency-detuned excitation. Iodine is chosen as the molecular system in which to evaluate the feasibility of these approaches due to its convenient visible absorption spectrum.

It is shown theoretically that complete saturation would eliminate the quenching dependence of the fluorescence signal. Saturation experiments are performed which indicate that with available continuous-wave laser sources of Gaussian transverse intensity distribution only partial saturation can be achieved in iodine at the pressures of interest in gasdynamics. Therefore, it is concluded that saturation is not a viable approach to eliminating the quenching complication.

Using a fluorescence lineshape theory it is shown that for sufficiently large detuning of a narrow bandwidth laser from a molecular transition

the quenching can be cancelled by collisional broadening over a large range of pressures and temperatures. Experimental data are obtained in a room temperature static cell which allow determination of the molecular quenching, collisional broadening and effective hyperfine-width constants. Data are obtained in a Mach 4.3 underexpanded jet of nitrogen, seeded with iodine, for various single-mode argon laser detunings from a strong iodine transition at 5145 Å. Using the experimentally determined molecular constants and the lineshape theory, good agreement is shown between the experimental and calculated fluorescence distribution in the jet flowfield. For a detuning of 3 GHz the signal is seen to be proportional to the iodine density and independent of the gasdynamic pressure and temperature over most of the flowfield; thus, the quenching dependence is effectively suppressed. The conclusion is drawn that LIF becomes a quantitative density probe by frequency detuning a narrow bandwidth laser from the molecular transition.

2. J.C. McDaniel, D. Baganoff and R.L. Byer, "Density Measurement in Compressible Flows Using Off-Resonant Laser-Induced Fluorescence", The Physics of Fluids, Vol. 25, No. 7, p. 1105, July 1982.

#### Abstract

Measurement of molecular number density in compressible flows using laser-induced fluorescence is complicated by collisional quenching of the excited state. It is shown that by exciting the fluorescence off-resonance the signal becomes proportional to number density and independent of collisional effects. Quantitative measurements of density in an underexpanded jet of nitrogen is demonstrated using off-resonant

fluorescence from iodine seed molecules irradiated with the 514.5 nm line of the argon-ion laser.

## PRESENTATIONS

1. J.C. McDaniel and D. Baganoff, "Density Measurements Using Laser-Induced Fluorescence", 34th Annual Meeting of the Division of Fluid Dynamics, Monterey, CA, Nov. 1981.

### Abstract

Laser-Induced fluorescence is investigated as a quantitative density measurement scheme in compressible flows. A fluorescence lineshape theory is used to show that the complication of collisional quenching can be cancelled by collisional broadening for sufficiently large detuning of a narrow bandwidth laser. Data are collected in a static nitrogen cell and in a Mach 4 underexpanded nitrogen jet, both seeded with iodine, for various single mode argon laser detunings from a strong iodine transition. For a detuning of 3 GHz the data and calculations show that the fluorescence is proportional to the gasdynamic density in a premixed flow. The method greatly reduces the normal effect of collisional quenching which causes the fluorescence intensity to be related to the density in a complicated way. Photographs of a cross-section of the jet flowfield show the dramatic change in the fluorescence distribution caused by the detuning. The prospects of utilizing other laser sources for measurement of density distribution in a plane of an iodine-seeded compressible flowfield are reviewed.